In-Situ Hydrogen Reduction of Lunar Polar Regolith: from Proof of Concept Experiments with ProSPA to Larger Scale ISRU Demonstrators

Conference or Workshop Item

IN-SITU HYDROGEN REDUCTION OF LUNAR POLAR REGOLITH: FROM PROOF OF CONCEPT EXPERIMENTS WITH PROSPA TO LARGER SCALE ISRU DEMONSTRATORS. P. Reiss[1], L. Grill[1], H. Sargeant[2], and S. Barber[2], [1]Institute of Astronautics, Technical University of Munich, Boltzmannstr. 15, 85748 Garching, Germany, (p.reiss@tum.de), [2]The Open University, Milton Keynes, MK7 6AA, UK.

Introduction: The European Space Agency (ESA) currently develops the Package for Resource Observation and in-Situ Prospecting for Exploration, Commercial exploitation and Transportation (PROSPECT) as a payload contribution to the planned Russian Luna-27 mission to the lunar polar regions. The package contains the sample analysis instrument ProSPA (PROSPECT Sample Processing and Analysis) to investigate volatile compounds that are extracted by heating regolith samples delivered by a drill [1-4]. In support of the instrument development, the Institute of Astronautics at Technical University of Munich (TUM) has established a breadboard to study the capability to extract oxygen from lunar regolith for the demonstration of In-Situ Resource Utilisation (ISRU) with ProSPA. The chemical process applied for this purpose is the reduction of iron oxides in the lunar regolith using hydrogen as reducing agent. The product of this reaction is water, which in a later ISRU application could be used directly or split via electrolysis to derive oxygen and hydrogen. Similar studies have been performed at the Open University with a different breadboard [5].

Breadboard: The ProSPA laboratory breadboard at TUM allows the heating of a regolith analogue sample with 2.8 mm in diameter and 4.5 mm in height (33-36 mg at a density of 1.2-1.3 g/cm^3), according to the ProSPA baseline sample size, in a high vacuum (<10^{-5} mbar). The breadboard design is described in detail by [6-7], a cut view of the sample container and a flow diagram are provided in Figure 1 and 2. Because the prime purpose of ProSPA is the extraction of volatile compounds from lunar regolith, the breadboard is not optimised for hydrogen reduction. As a consequence, the regolith sample is not ideally exposed to the reactant gas. As visible from Figure 1 and 2, the sample is present as a fixed bed at the end of the vacuum tubing, while the reactant gas enters and leaves the fixed bed from the top. A concern with such a configuration is that not all of the sample particles are exposed to the reactant and that the produced water accumulates in the sample, preventing further reduction.

Process: To investigate if the chemical reduction can be realised despite these concerns, several different operational modes were evaluated with the breadboard, as described in the following.

(1) Continuous gas flow: The sample is constantly purged with hydrogen at an absolute pressure of ~10^1 mbar and heated at 800 °C for 3 h. The gas mixture is permanently analysed by the RGA.

(2) Semibatch with preheating: Hydrogen is introduced into the sample compartment at an absolute pressure of ~10^3 mbar and the sample is heated to 800 °C for 30 min. Subsequently, the dosing valve is opened and gas is leaked to the RGA for analysis.

(3) Semibatch without preheating: Hydrogen is introduced into the sample compartment at an absolute pressure of ~10^3 mbar. The sample is heated to 800 °C and the dosing valve opened for gas analysis at the same time.

For all modes, the samples were baked-out prior to test to remove volatiles and atmospheric contamination. The samples used in the experiments were the lunar highland type regolith simulant NU-LHT-2M with standard particle size distribution and 93-96 % pure ilmenite ($FeTiO_3$) with a mean particle size of 174 µm. Additionally, a third run without sample was performed for each operational mode for reference.

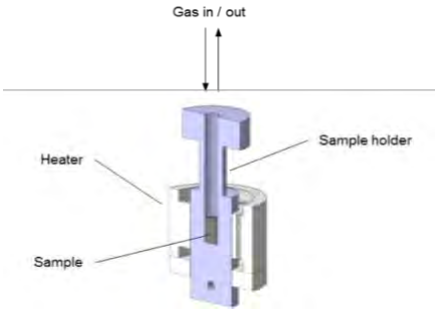

Figure 1: Cut view of the sample container

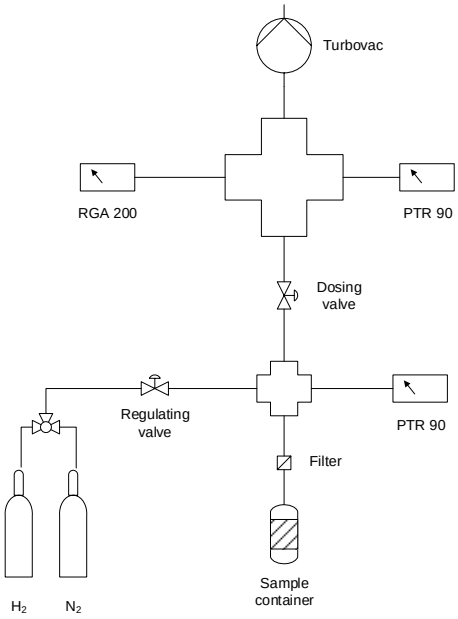

Figure 2: Flow diagram of the breadboard

Test Results: While for the first two operational modes no measurable production of water was detected, the third operational mode (semibatch without preheating) lead to a distinct raise in the partial pressure of water with the ilmenite sample. In the first 30 min of heating, the ratio of partial pressures of m/z 18 and m/z 2 (ratio of product to reactant) rises from 0.5 % to 10 % (Figure 3). For NU-LHT-2M, no significant change of this ratio was detected, comparable to the reference run without sample. An estimation of the mass of water vapour produced during the reaction, based on the temporal integration of the partial pressure, yielded values up to ~2 wt% for the ilmenite sample after a process duration of 3 h.

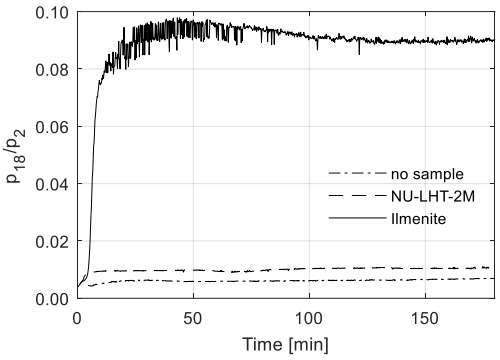

Figure 3: Partial pressure ratio water/hydrogen for the semibatch mode without preheating

Process Evaluation: The production of water from lunar regolith and its analogues is mainly done through reduction of FeO, but also TiO_2 and SiO_2. A major source of FeO that is susceptible to reduction by hydrogen is ilmenite. Lunar highland regolith typically has very low FeO and TiO_2 content (see Table 1), compared to mare regolith with FeO contents from 10-22 wt% and TiO_2 contents from 2-12 wt% [8]. As studied by [9], the oxygen yield of the present chemical reduction depends directly on the iron content: For the Apollo highland sample 62241, an oxygen yield of just over 1 % was achieved, while for the mare soils 12032 and 71131, 1.5 % and 3.5 % oxygen were achieved respectively.

Considering the above, it was expected that NU-LHT-2M would not yield a significant amount of water in the discussed experiments. However, it was not fully determined whether this was only due to low FeO content or if the non-optimised oven design has a major influence on the process efficiency. A possible risk is that the reactant gas does not diffuse through the entire sample leading to decreasing degrees of reduction over sample depth. Other risks include the resorption or reconversion of produced water within the sample.

Outlook: Based on the proof of concept studies with the ProSPA setup, further investigations will be made that will also inform the development of scaled-up ISRU demonstrators and dedicated ISRU payloads in the near future.

ProSPA breadboard. Ongoing trade studies on the effect of several process parameters on the efficiency of the reduction are being addressed. This mainly includes the reactant gas pressure, sample temperatures, and sample type and particle size. Additional tests will be conducted to study the gas permeability of the sample and the potential of resorption or reconversion of the produced water before it leaves the sample. An improved version of the sample oven will be evaluated to assess how the current design can be optimised for ISRU purposes, for example that the reactant gas is flown through the sample.

ISRU demonstrator. The reduction of lunar regolith with hydrogen has a relatively low efficiency with oxygen yields around 2-3 wt% from pure ilmenite. The major objective for the design of future demonstrators therefore is to increase the process efficiency. Instead of using a static fixed-bed reactor such as for ProSPA or other laboratory studies [11,12], the best way of improving the reaction is to establish a continuous gas flow through the sample. This would enable better exposure of the sample to the reactant and ensure that the produced water is transported out of the sample to keep the reaction running. With larger sample quantities it is beneficial to add further sample agitation methods, such as a rotated/oscillating bed, a mixing auger [13,14], or a fluidized bed [15].

Table 1: Major oxides in Apollo 16 regolith and NU-LHT-2M (average and standard deviation in wt%)

Oxide	Apollo 16 soils and breccia [8]		NU-LHT-2M certificate of information		NU-LHT-2M material safety datasheet [10]	
	Avg.	SD	Avg.	SD	Avg.	SD
SiO_2	44.89	0.51	47.62	0.21	46.70	0.11
Al_2O_3	27.23	1.84	27.4	0.48	24.40	0.07
CaO	14.56	3.64	13.6	0.41	13.60	0.05
MgO	6.00	1.76	8.50	0.48	7.90	0.04
FeO	4.98	1.08	2.15	0.09	-	-
Fe_2O_3	-	-	-	-	4.16	0.03
TiO_2	0.53	0.14	0.33	0.02	0.41	0.02
Na_2O	0.47	0.08	1.43	0.03	1.26	0.02

References: [1] Barber S. J. et al. (2017) *ELS*. [2] Barber S. J. et al. (2017) *LPSC*. [3] Carpenter J. et al. (2014) *LEAG*. [4] Fisackerly R. et al. (2015) *ASTRA*. [5] Reiss P. (2017), *ELS*. [6] Sargeant H. et al (2017) *ELS*. [7] Reiss P. (2018), Dissertation, *TUM*. [8] Haskin L. and Warren P. (1991), *Lun. Sourcebook*. [9] Allen C. C. (1997), *NASA Lunar News No. 60*. [10] U.S. Geological Survey (2008), *NU-LHT-2M MSDS*. [11] Taylor L. et al. (1993), *LPSC*. [12] Yoshida H. et al (2000), 2nd Space Res. Roundtable. [13] Lee K. A. et al. (2013), *J. Aerosp. Eng.*, 26(1), 67–73. [14] Clark D. L. et al. (2009), *AIAA Sp. Conf. Expo*. [15] Taylor, L. and Carrier, W. D. (1993) *Res. of Near Earth Space*.